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Particle size distribution measurements using polarized light methods

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Abstract : In this paper, we pay attention to particle size distribution (PSD) measurements analyzing the polarization ratio of the light scattered by a polydisperse solution. The sample solutions were prepared mixing latex isotropic spheres of different sizes in order to simulate a variety of size distributions. Mie scattering computations have been performed to fit the experimental data and thus to determine the sample PSD. Measurements were made using an optical bench-mounted ellipsometer employing a laser diode source as light source.

Keywords : Particle size distribu[illegible] pola[illegible] polydisp[illegible] solution

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Measurements employing polarized light have found a wide range of applications in industrial and medical environments [1-3].

Light scattering offers advantages over other techniques for particle analysis because the system under study can be observed *in situ* without significant perturbation. The method is absolute in the sense that the theory permits reduction of the data directly to the final desired results without the need of secondary schemes for calibration. In the particle size analysis, it is usually assumed that the scattering by an array of particles is incoherent so that the scattering functions corresponding to an isolated particle may be used and also there is no multiple scattering. The cumulative effect is obtained by adding the intensity scattered by each particle as if it were alone [2].

The most convenient suspensions for light scattering studies are the polymer latexes since they consist of isotropic spheres with very narrow distribution sizes. Furthermore, various sizes can be prepared, which can then be mixed to simulate a variety of size distributions. A normal distribution cannot represent a distribution of particle sizes with highest precision because it admits negative value of particle size. In addition, unlike the normal distribution which is symmetrical, naturally occurring populations are frequently skewed. A good representation of many populations is the logarithmic normal distribution that was used by us for computations and experimental determinations.

This method is based on the polarization measurements of the scattered radiation obtained with monochromatic light at various angles of observation. The intensity of the components of the scattered light whose electric vector vibrates perpendicular and parallel to the plane of observation, $I_1(\theta)$ and $I_2(\theta)$, is measured at different angles.

Relation (1) describes the polarization ratio

$$p(\theta) = \frac{I_2(\theta)}{I_1(\theta)} = \int p(\alpha) i_2(\theta,\alpha) d\alpha \Big/ \int p(\alpha) i_1(\theta,\alpha) d\alpha . \quad (1)$$

This being a ratio, it is sufficient to use instrument readings rather than absolute intensities. Here $p(\alpha)$ is the logarithmic normal distribution, sometimes called zeroth order logarithmic distribution (ZOLD) which is defined as

$$p(a) = \exp\left[-\frac{(\log a - \log a_M)^2}{2\sigma_0^2}\right] \Big/ (2\pi)^{1/2} \sigma_0 a_M \exp\left[\sigma_0^2/2\right]. \quad (2)$$

where a_M is the modal value of a and σ_0 is a measure of the width of the distribution. The relation between a and α is given by formula $a = \lambda\alpha/2\pi$. Thus, in order to measure the PSD, it is enough to determine α_M (or a_M) and σ_0 from the experimental data. This is done by comparison of the experimental values of $\rho(\theta)$ with theoretical computations corresponding to the refractive index of the system at the wavelength under investigation.

For any iterative matching technique, one must raise the question of the uniqueness of the solution. It was proved that between experimental and calculated results, the concordance is excellent. In general, a unique solution was obtained [2].

Most commercial turbidimeters use light sources, which emit unpolarized light. Laser diodes due to their characteristics (small size, low cost and high output intensity) have been suitable for turbidity measurements with polarized light. In addition, the light emitted by these devices is usually linear-polarized.

We used a slightly modified ellipsometer, with a laser diode as light source, for turbidity measurements (nephelometer). This kind of experimental arrangement has the advantage that one can modify both the scattering angle and also one can select a particular polarization state [4].

The sample containers were identical cylindrical bottles made of transparent soda lime-silica glass [4]. Each bottle was tested and the overall conclusion drawn was that the birefringence of the glass bottles was negligible.

The sample solution was obtained by mixing fourteen monodisperse latexes in order to approximate a ZOLD distribution given by relation (2). The histogram of this mixture was determined by electron microscopy and is presented in Figure 1.

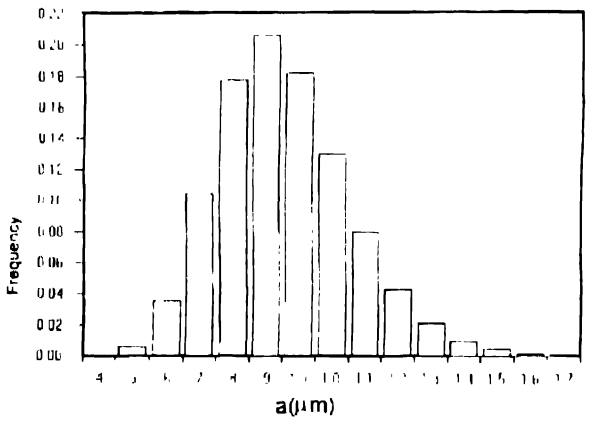

Figure 1. Electron microscopy histogram for latex mixture with a_M = 10 and σ_0 = 0.19

The theoretical ZOLD distribution curve computed using relation (2) is plotted in Figure 2.

The polarization ratio $\rho(\theta)$ was measured in the angle range between 30° and 130° with a step of 10°. Furthermore, we have computed theoretical polarization ratio $\rho(\theta)$ which

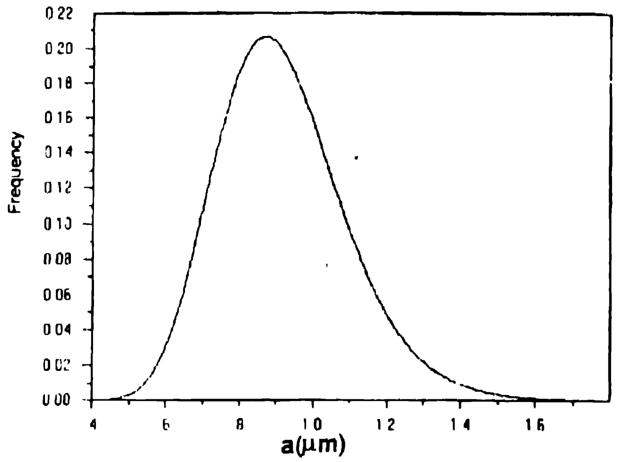

Figure 2. The theoretical ZOLD distribution curve with a_M = 10 and σ_0 = 0.19

fits best the experimental data [2]. For Mie scattering computing, we used an algorithm based on Dave's downward recurrence scheme [5–7]. The required mathematical functions are implemented in shared library that can be called from a relational database. Thus, one has the possibility to work in a multi-user, parallel processing and distributed environment and to store the computation results in the database. The mathematical functions can be used from the users on the LAN (local area network), WAN (wide area network) and on Internet through RPC (remote procedure call).

The results are plotted in Figure 3.

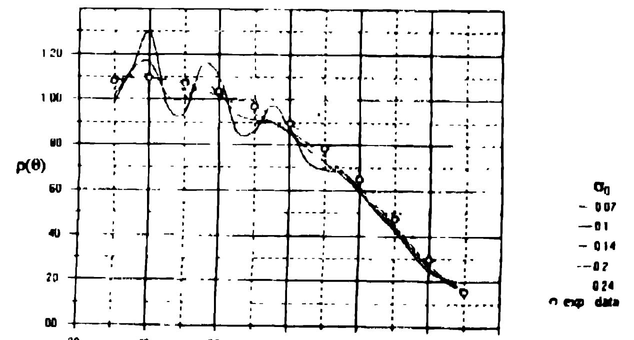

Figure 3. The experimental (circle) and theoretical plot (line) for polarization ratio $\rho(\theta)$ with a_M = 10. The best fit between experience and computational data is obtained for σ_0 parameter equal to 0.2

In Figure 3, it is performed a tuning for σ_0 parameter and it is obvious that the best fit is obtained when $\sigma_0 = 0.2$. The result is in very good agreement with experimental values.

In addition, we have tested the $\rho(\theta)$ parameter variation with a_M (Figure 4). It is obvious that the best fit is obtained for a_M = 10 and the solution uniqueness is guaranteed.

Taking into account the required computational effort in order to find out the right PSD, we tried to reduce the amount of intricate Mie scattering computations. Consequently, we

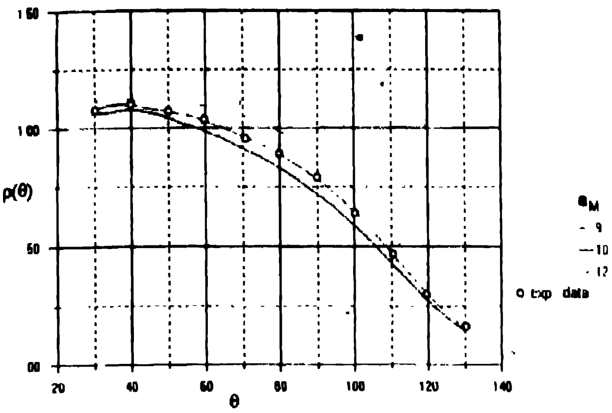

Figure 4. The experimental (circle) and theoretical plot (line) for polarization ratio $\rho(\theta)$ with $\sigma_0 = 0.2$ The best fit between experience and computational data is obtained for a_M parameter equal to 10

have computed the polarization ratio $\rho(\theta)$ for entire expected range of (a_M, σ_0) parameters and then we have stored the results into a relational database. Furthermore, the right PSD determination was accomplished easily by performing a query against database and retrieving (a_M, σ_0) parameters that fit the experimental $\rho(\theta)$ angular distribution best. Thus, it is not necessary to make complex Mie scattering computations every time when experimental measurements are performed and the PSD is obtained instantly

Even if the method described above is known for more than thirty years [2], it has also the advantage that does not require complex experimental arrangements and expensive technical means. It has also the advantage that combined with a relational database (as we have described above), the method can be used to provide very good results for a large variety of turbidity measurements involving emulsions without further computational effort.

The drawback of the method is the fact that we need *a priori* knowledge of the PSD shape in order to performed the Mie scattering computations.

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